Capillary wrinkling scaling laws of floating elastic thin films under a drop

Bohua Sun[1]

[1] *Cape Peninsula University of Technology, Cape Town, South Africa*
sunb@cput.ac.za

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This paper uses dimensional analysis to define the general expression of the pair (N, ℓ), and identifies the dominant combination-parameters of the capillary wrinkling problem, while it also determines the dominant parameters of different problems relating to its use. The dimensional analysis results reveal that, in general, there are no universal scaling laws for capillary wrinkling. Only for a small/moderate deformation, it was found that the wrinkling number N is mainly controlled by the ratio of bending stiffness and surface tension, while the wrinkling length ℓ is controlled by the ratio of in-plane stiffness and surface tension. Having linear physical relationship in the case of the small deformation, simpler scaling laws are proposed for the pair (N, ℓ). The universality of the scaling laws, which are verified by the dimensional analysis, will give us more confidence. As a natural extension, we gave the pair (N, ℓ) a thin film case made of axisymmetric anisotropic materials. By using Tanner's scaling laws, we obtained dynamical scaling laws for a drop radius and the pair (N, ℓ), which shows that the pair (N, ℓ) will fade away with time. Finally, we obtained the pair (N, ℓ) within the gravity regime.

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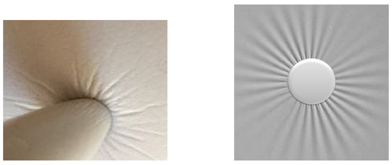
INTRODUCTION

The deformation patterns of elastic membranes under tension is called wrinkling. Wrinkling, which is caused by capillary surface tension, is called capillary wrinkling (Figure 1).



(a) (b)

FIG. 1: a. Typical wrinkling of elastic film; b. Typical capillary wrinkling of elastic film. The problem is to find the deformation wrinkling pattern pair (N, ℓ), where the wrinkling number is N and the wrinkling length is ℓ

In recent years wrinkling patterns have drawn particular attention [1–11], since it can be a useful tool to infer material parameters that might otherwise be inaccessible. For example, the commonly observed tearing instability of an elastic sheet, adhered to a rigid substrate, can also be used to characterize the adhesion energy, which is the traction forces that are exerted by fibroblasts during cell division to the mechanical properties of the membranes themselves (such as Young's modulus and thickness) [5].

There has also been considerable interest in understanding fundamental aspects of wrinkled membranes including wrinkling of a compressed elastic film on a viscous layer [1], wrinkling mechanics and the geometry of an elastic sheet under tension [2, 3], the size and number of wrinkles [5], the transition from wrinkling to folding [6], the analytic analysis of capillary wrinkling of the circular elastic membranes [7], wrinkling of pressurized elastic shells [8], wrinkling of a charged elastic film on a viscous layer [9] and capillary deformations of bendable films [10].

Regarding the capillary wrinkling of thin film, a milestone has been laid by Huang *et al.* [5] and Vella *et al.* [7]. Huang *et al.* [5] experimentally derived at the length and numbers of capillary wrinkling, while Vella *et al.* [7] theoretically proved the capillary wrinkling length expression obtained by Huang *et al.* [5] for a small deformation of a circular film. A novel experiment, combining both fundamental and applied aspects of the interaction between surface tension and elasticity, was presented by Huang *et al.* [5]. In this experiment, a small liquid drop was placed onto an elastic membrane that floated in a bath of the same liquid. Before adding the drop, the membrane was stretched by the surface tension of the liquid bath. Once the drop was added, owing to the opposing tension, the contact line of the drop caused radial wrinkles. Experimentally, it was found that the wrinkling length $\ell = 0.031R(\frac{Eh}{\gamma})^{1/2}$, where r is the radius of the drop and γ is the surface tension coefficient of the liquid-gas interface. This pure empirical relationship is confirmed by a theoretical justification of Vella *et al.* [7], who used the Föppl-von Kárman thin plate theory.

Although Huang *et al.* [5] obtained the formula for the pair (N, ℓ), where N was wrinkling number, the question remains are these relationships universally applicable? If the general situation is not, then under what circumstances can it be approximated to the universal scale law? How can one promote other materials such as axisymmetric anisotropic materials? The obtained pair (N, ℓ) was the static result, so how is the scale law of the

dynamics problem established? If the characteristic scale of the drop is greater than the capillary scale κ^{-1}, gravity begins to dominate the wrinkling process, so what is the scale laws beyond the capillary scale κ^{-1}?

The paper first introduce the research topic before presenting the general expression of the pair (N, ℓ) by using dimensional analysis and by determining the controlling parameters of the problem,while proposing linear approximated scaling laws for the pair based on the understanding of the test's data, and then extending to the film made of composite and laminated materials. For the spreading of a drop, dynamics of spreading scaling laws are proposed based on Tanner's law. Finally, the paper concludes with a formulation of wrinkling scaling laws in the gravity regime.

CAPILLARY WRINKLING SCALING LAWS FOR SMALL/MODERATE DEFORMATION

Generally speaking, both the wrinkling number and length are functions of the Young modulus E, film thickness h, radius of drop r and surface tension γ, namely, $N = F(E, h, \nu, \gamma, r)$ and $\ell = G(E, h, \nu, \gamma, r)$, where the F and G are unknown functions. The dimensions of those parameters are listed in Table I below.

TABLE I: Parameters and Dimensions

N	ℓ	h	E	ν	γ	r
1	L	L	$\mathrm{L^{-1}MT^{-2}}$	1	$\mathrm{MT^{-2}}$	L

Since the film will be in a bending and stretching state by dimensional analysis [12–14], the above formula can be further expressed into the following forms: $N = F(D, K, \gamma, r)$ and $\ell = G(D, K, \gamma, r)$, where the bending stiffness $D = Eh^3/[12(1-\nu^2)]$ and the in-plane stiffness $K = Eh/(1-\nu^2)$. They can also be expressed in dimensionless format, as follows:

$$N = F(r\sqrt{\frac{\gamma}{D}}, \sqrt{\frac{K}{\gamma}}), \tag{1}$$

$$\frac{\ell}{r} = G(r\sqrt{\frac{\gamma}{D}}, \sqrt{\frac{K}{\gamma}}). \tag{2}$$

These are the general scaling relations for capillary wrinkling of the floating thin film. However, both functions F and G were not able to be confirmed by the dimensional analysis, before other methods must be used. These relations reveal that the pair (N, ℓ) will be controlled by two dimensionless parameters, namely $r\sqrt{\gamma/D}$ and $\sqrt{K/\gamma}$.

Regarding the capillary wrinkling of elastic film or membranes, Huang *et al.* [5] conducted a good test. From the test data we found that the wrinkling number N mainly depends on the bending stiffness D; however, the wrinkling length ℓ mainly depends on the in-plane stiffness K. Therefore, Eqs. (1) and (2) can be simplified as follows:

$$N = F(r\sqrt{\frac{\gamma}{D}}), \tag{3}$$

$$\frac{\ell}{r} = G(\sqrt{\frac{K}{\gamma}}). \tag{4}$$

Theoretically, these are the relations that we obtain from dimensional analysis. General speaking, the functions F and G are not in power forms, which means that capillary wrinkling phenomena has no power laws and/or scaling laws. Nevertheless, these relations can still provide some useful information such as the fact that dimensionless parameter $r\sqrt{\gamma/D}$ is a control variable for the wrinkling number N; while the parameter K/γ will be a control variable for the wrinkling length ℓ.

For a small deformation physical laws must be linear; therefore, Eqs. (3) and (4) should be a linear function of the controlling parameters as shown below

$$N \approx \left[C_N \left(r\sqrt{\frac{\gamma}{D}}\right)^{\alpha}\right], \tag{5}$$

$$\frac{\ell}{r} \approx C_\ell \left(\sqrt{\frac{K}{\gamma}}\right)^{\beta}, \tag{6}$$

where $[Y]$ represents the integer part of real number Y, while the constants C_N, C_ℓ and exponents α, β can be determined by either numerical simulations or experiments.

For a small deformation from the data fitting, Huang *et al.* [5] found $C_r = 0.033$, $C_\ell = 3.62$ and $\alpha = 1/2$, $\beta = 1/2$. Hence, the scaling laws are as follows

$$\frac{\ell}{r} = 0.033(\frac{K}{\gamma})^{1/2}, \tag{7}$$

and the number of wrinkling N

$$N = \left[3.62\left(r\sqrt{\frac{\gamma}{D}}\right)^{1/2}\right]. \tag{8}$$

It is worth pointing out that Eqs. (7) and (8) are universal and are valid for all thin flat film that have a small deformation. The scaling laws in Eqs. (7) and (8) can be used to facilitate measurements of the bending stiffness and in-plane stiffness K. If you can firstly get the N, ℓ and radius r, hence, we can calibrate the D and K as follows

$$D = \frac{3.62}{N}\gamma\sqrt{r}, \tag{9}$$

$$K = 918.27\gamma(\frac{\ell}{r})^2. \tag{10}$$

and the film thickness h

$$h = \sqrt{\frac{12D}{K}} = \frac{1}{100}N^{-1/2}\ell^{-1}r^{5/4}. \tag{11}$$

These scaling laws have been theoretically confirmed by Vella *et al.* [7], who used von Kárman's circular plate theory and hence derived at the similar scaling laws of capillary wrinkling.

With the help of Eqs. (3) and (4), the above scaling laws of N and ℓ can be extended to the film, which comprises composites materials and laminated materials by simply replacing D and K with a corresponding e-quivalent or effective bending stiffness D_{eff} and in-plane stiffness K_{eff}; hence,

$$N = \left[3.62\left(r\sqrt{\frac{\gamma}{D_{eff}}}\right)^{1/2}\right], \qquad (12)$$

$$\frac{\ell}{r} = 0.033(\frac{K_{eff}}{\gamma})^{1/2}. \qquad (13)$$

These expressions provide a pretty accurate estimation for those materials without further investigation. This is one of advantages of having universal scaling laws that are verified by dimensional analysis, which definitely gives us ultimate confidence in such an extension.

For example, if the thin film is made of axial symmetric orthogonal materials, $D_{eff} = D_r$ and $K_{eff} = K_r$, we have the pair $N = \left[3.62\left(r\sqrt{\gamma/D_r}\right)^{1/2}\right]$, and $\frac{\ell}{r} = 0.033(K_r/\gamma)^{1/2}$, with the equivalent/effective bending stiffness $D_r = E_r h^3/[12(1-\nu_r\nu_\theta)]$ and the equivalent/effective in-plane stiffness $K_r = E_r h/(1-\nu_r\nu_\theta)$, radius Young modulus E_r and the Poisson ratio ν_r, and the circle Poisson ratio ν_r.

CAPILLARY WRINKLING DYNAMICS OF A TOTALLY WETTABLE FILM

Static wrinkling problem has been investigated, however, no dynamical wrinkling has been studied yet. A drop on a film surface, within a complete wetting regime will slowly spread. Typically, the spreading lasts from a few hours for ordinary liquids, to several weeks for highly viscous fluids such as heavy silicone oils (Figure 2).

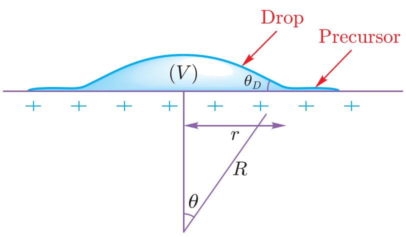


FIG. 2: Spreading of a drop on a film surface in a total wetting regime. Tanner's law: $\theta_D \sim t^{-3/10}$. In this paper, we obtained $r \sim t^{-3/5}$

This dynamic process can be expressed in terms of a contact angle θ_D, which depends on the spreading time t. When surfaces are smooth and clean, and for non-volatile liquids, Tanner [16] obtained a remarkable universal law: $\theta_D \sim t^{-3/10}$.

The measurements reveal a highly surprising fact, namely that the angle θ_D is completely independent of the spreading parameter $S = \gamma_{SO} - \gamma_{SL} - \gamma$ as long as S is positive, that is to say, as long as we are in a total wetting regime, where the three coefficients γ are the surface tensions at the solid/air, solid/liquid, and liquid/air interfacer, respectively. This is surprising because the force F that acts on the system of interest is essentially equal to spreading parameter $F = \gamma_{SO} - \gamma_{SL} - \gamma\cos\theta_D \approx \gamma_{SO} - \gamma_{SL} - \gamma = S$. There are two wetting regimes for sessile drops. Partial wetting ($S < 0$): The drop does not spread, but instead forms a spherical cap at equilibrium, resting on the substrate with a contact angle θ_D. A liquid is said to be "mostly wetting" when $\theta_D < \pi/2$, and "mostly non-wetting" when $\theta_D > \pi/2$. Total wetting ($S > 0$) : if the parameter S is positive, the liquid spreads completely in order to lower its surface ($\theta_D = 0$). The Law of Young-Dupr'e: $\gamma\cos\theta_D = \gamma_{SO} - \gamma_{SL}$, and $S = \gamma(\cos\theta_D - 1)$.

The precursor film is evidence of the great force F that acts on its boundary. The liquid is rapidly drawn towards the periphery in the form of a film whose thickness is roughly a pancake's thickness, and which is defined as $e = 2\kappa^{-1}\sin(\theta_D/2)$ as shown in Figure 3 below.

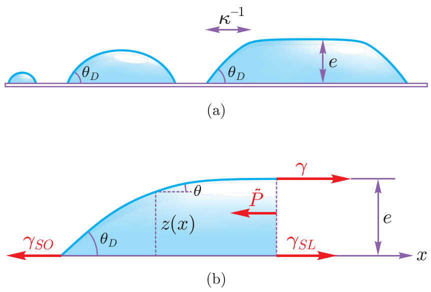


FIG. 3: (a) Liquid drops of increasing size on a sheet of film. Gravity causes the largest drops to flatten. (b) Equilibrium of the forces (per unit length of the line of contact) act on the edge of a puddle. $\tilde{P} = (1/2)\rho g e^2 = -S$ is the hydrostatic pressure. The equilibrium of forces that act on the line of contact, $\gamma(1-\cos\theta_D) = (1/2)\rho g e^2$, gives the thickness $e = 2\kappa^{-1}\sin(\theta_D/2)$, where the capillary length, $\kappa^{-1} = \sqrt{\gamma/(\rho g)}$

But, behind the film the forces that are involved are quite different. Within the drop are forces of traction $-\gamma_{SL} - \gamma\cos\theta_D$, whereas within the film (characterized by a zero angle) there are proper forces $\gamma_{SL} + \gamma$. The net force that acts on the drop is then only $F = \gamma(1-\cos\theta_D) \approx \frac{1}{2}\gamma\theta_D^2$, velocity $V = (V^*/6l)\theta_D^3$, where the dimensionless coefficient $15 < l < 20$, $V^* = \dot{\gamma}/\eta$, and viscosity η. From conservation of the volume $\Omega = (\pi/4)R^3\theta_D$ of the drop, it is easy to obtain the angle $\theta_D \approx (\Omega^{1/3}/V^*)^{3/10}t^{-3/10}$, and the radius $R(t) \approx \Omega^{1/3}(V^*/\Omega^{1/3})^{1/10}t^{1/10}$.

Therefore, the radius of the drop is given by

$$r \approx e \tan\theta_D \approx \kappa^{-1}\theta_D^2 = \kappa^{-1}(\frac{\Omega^{1/3}}{V^*})^{3/5}t^{-3/5}. \quad (14)$$

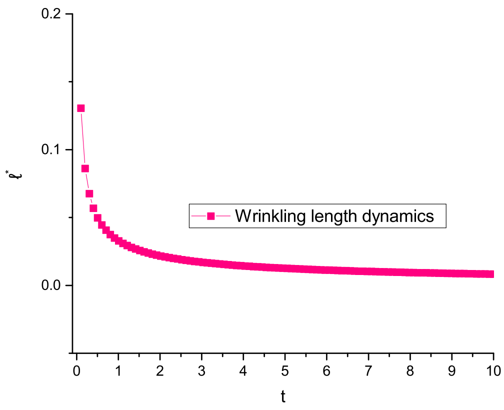
This scaling laws of the radius of a drop has not been recorded in literature. When one substitutes this into Eqs(5,6), one has the wrinkling length

$$\begin{aligned} \ell &= 0.033(\tfrac{K}{\gamma})^{1/2}\kappa^{-1}(\tfrac{\Omega^{1/3}}{V^*})^{3/5}t^{-3/5} \\ &= 0.033(\tfrac{K}{\gamma})^{1/2}\kappa^{-1}(\tfrac{\eta\Omega^{1/3}}{\dot{\gamma}})^{3/5}t^{-3/5}, \end{aligned} \quad (15)$$

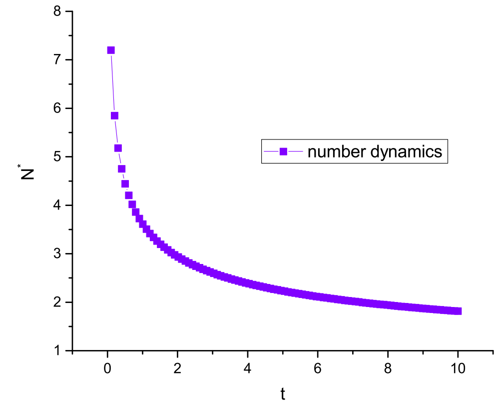
and the wrinkling number

$$\begin{aligned} N &= [3.62(\tfrac{\gamma}{D})^{1/4}\sqrt{\kappa^{-1}}(\tfrac{\Omega^{1/3}}{V^*})^{3/10}t^{-3/10}] \\ &= \left[3.62(\tfrac{\gamma}{D})^{1/4}\sqrt{\kappa^{-1}}(\tfrac{\eta\Omega^{1/3}}{\dot{\gamma}})^{3/10}t^{-3/10}\right]. \end{aligned} \quad (16)$$

The dynamics of both length and number are illustrated in Figure 4.



(a)



(b)

FIG. 4: (a) Capillary wrinkling length dynamics $\ell^* = \ell/\left[(\frac{K}{\gamma})^{1/2}\kappa^{-1}(\frac{\Omega^{1/3}}{V^*})^{3/5}\right]$; (b) Capillary wrinkling number dynamics $N^* = N/\left[(\frac{\gamma}{D})^{1/4}\sqrt{\kappa^{-1}}(\frac{\eta\Omega^{1/3}}{\dot{\gamma}})^{3/10}\right]$

CAPILLARY WRINKLING WITHIN THE GRAVITY REGIME

From Eqs.(14,15), it is interesting to note that both ℓ and N fade away with spreading time, and will stop at critical time $t_c = \Omega^{1/3}/V^* = \eta\Omega^{1/3}/\dot{\gamma}$. Beyond the critical time, the spreading enters the gravity regime. It is important to bear in mind that this equation applies only when r is less than the capillary length κ^{-1} . When $r > \kappa^{-1}$, gravity must be taken into account [15].

There exists a particular length, denoted $\kappa^{-1} = \sqrt{\gamma/(\rho g)}$, beyond, which gravity becomes important, and is referred to as the capillary length. The length κ^{-1} is generally of the order of a few mm. If one wants to increase the length κ^{-1}, it is necessary to work in a microgravity environment or, more simply, to replace air with a non-miscible liquid whose density is similar to that of the original liquid [15].

Gravity is negligible for sizes $r < \kappa^{-1}$. When this condition is met, it is as though the liquid is in a zero-gravity environment and capillary effects dominate. The opposite case, when $r > \kappa^{-1}$, is referred to as the "gravity" regime.

In the critical situation $r = \kappa^{-1}$ and beyond, the scaling laws Eq.(12) of the wrinkling length ℓ will be independent of the surface tension γ as follows

$$\ell = 0.033(\frac{K}{\rho g})^{1/2}, \quad (17)$$

and the wrinkling number N, given by

$$N = \left[\frac{3.62\gamma}{\sqrt{\rho g D}}\right]. \quad (18)$$

It is clear that both N and ℓ are constant within the gravity regime.

CONCLUSION

This paper has attempted to answer all the previous questions. In general, there are no universal scaling laws for capillary wrinkling. Only in the case of small and moderate deformation, can special universal scaling laws be formulated. Regarding the bending and in-plane stiffness, it was found that the wrinkling number N is mainly controlled by the ratio of bending stiffness and surface tension, and the wrinkling length ℓ is controlled by the ratio of in-plane stiffness and surface tension. By using Tanner's scaling laws, we obtained dynamic scaling laws for a drop radius and the pair (N, ℓ), which shows that the pair (N, ℓ) will fade away with the time. Finally, the pair (N, ℓ) within the gravity regime was also revealed.

In summary, the highlights of this paper are listed in the Table II and III:

TABLE II: Static capillary wrinkling pair (N, ℓ)

Dry surface (Surface tension dominate)	Wet surface (Gravity dominate)
$N = [3.62\left(r\sqrt{\frac{\gamma}{D}}\right)^{1/2}]$	$N = [\frac{3.62\gamma}{\sqrt{\rho g D}}]$
$\ell = 0.033r(\frac{K}{\gamma})^{1/2}$	$\ell = 0.033(\frac{K}{\rho g})^{1/2}$

TABLE III: Dynamic capillary wrinkling pair (N, ℓ)

$N = [3.62(\frac{\gamma}{D})^{1/4}\sqrt{\kappa^{-1}}(\frac{\eta\Omega^{1/3}}{\dot{\gamma}})^{3/10}t^{-3/10}]$
$\ell = 0.033(\frac{K}{\gamma})^{1/2}\kappa^{-1}(\frac{\eta\Omega^{1/3}}{\dot{\gamma}})^{3/5}t^{-3/5}$

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